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FABRICATION OF THIN TUNGSTEN – URANIUM DIOXIDE COMPOSITE PLATES

by Gordon K. Watson
Lewis Research Center
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FABRICATION OF THIN TUNGSTEN - URANIUM DIOXIDE COMPOSITE PLATES*

by Gordon K. Watson

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SUMMARY

Various parameters affecting the fabrication of thin plates containing 20 volume percent uranium dioxide (UO_2) dispersed in a tungsten matrix have been investigated, and a standard method of fabrication by a powder-metallurgy technique and hot rolling has been developed. The effect of different fuel loadings (10 to 40 volume percent UO_2) on fabrication has been studied using -270/+400 mesh (53 to 37 μ) spherical fuel particles. A method of roll bonding wrought tungsten foil to major surfaces of the fuel plates to serve as a cladding material has also been developed.

The results of this investigation show that dense tungsten - uranium dioxide (W-UO_2) plates containing 10 to 40 volume percent UO_2 can be successfully fabricated using the procedure described in this report, and that the surfaces of the fuel plates can be clad with a thin, dense, uniform layer of unalloyed tungsten. The flat W-UO_2 plates can then be hot formed into cylindrical fuel-element configurations.

INTRODUCTION

The nuclear rocket program has created interest in metallic-base fuel elements having very high temperature capabilities (4000° F or better). Dispersion-type fuel elements in which the fissile phase is dispersed in a suitable metal matrix are of prime importance. Tungsten is of interest as the matrix material for this type of fuel element because of its high-temperature strength (ref. 1) and its compatibility with the hydrogen propellant. Uranium dioxide (UO_2) is of interest as the fissile material because it is one of the most refractory of the uranium compounds (mp, ~5000° F), because it has a high uranium content, and because it is relatively compatible with both tungsten (ref. 2) and hydrogen.

A nuclear rocket concept under consideration at the Lewis Research Center uses a water-moderated thermal reactor as a heat source for the hydrogen propellant (ref. 3). The nuclear characteristics of this thermal reactor permit the use of fuel elements containing relatively low percentages of fuel material, that is, less than 40 volume percent UO_2 dispersed in tungsten. This

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low fuel loading is advantageous from the materials aspect since it allows the elements to have properties approaching those of the metal matrix rather than those of the ceramic fuel. A fuel element being considered in one of the current reactor design concepts is an assembly of thin-walled concentric cylinders. This configuration provides a high ratio of surface area to volume and thus promotes efficient heat transfer from the fuel element to the propellant. Preliminary design studied have indicated that the fuel elements should contain fuel loadings in the range of 10 to 30 volume percent UO_2. Initial fabrication studies at the Lewis Research Center have concentrated on composites with a midrange loading of 20 volume percent fuel.

Previous work at the Lewis Research Center (refs. 4 and 5) established the feasibility of producing thin fuel plates containing UO_2 in a tungsten matrix by a powder-metallurgy and hot-rolling technique. The previous work also indicated that the surfaces of the fuel plates must be clad with a dense layer of unfueled tungsten to minimize the loss of UO_2 at elevated temperatures.

The purposes of this current investigation were to develop and optimize the fabrication technique for producing thin tungsten - uranium dioxide ($W-UO_2$) composites (approximately 0.020 in. thick) and to determine the effect of different fuel loadings on the fabrication of the plates. Part of the program was aimed at developing a simple cladding method that would produce a dense uniform clad of controlled thickness. In addition, preliminary studies on the forming of the flat fuel plates into various geometries were made. Results of these studies are described in this report.

MATERIALS

Five commercial tungsten powders of different particle size (0.88, 1.15, 2.1, 3.5, and 4.5 μ average particle size) were used in the initial phases of this investigation. The particle size distribution for these powders is given in table I. Tungsten powder, having an average particle size of 0.88 micron, was used in all subsequent phases of the study. Table II gives the chemical analysis of this powder. Commercial, wrought tungsten foil (0.003 in. thick) having a minimum purity of 99.9 percent was used as cladding for the fuel plates.

TABLE I. - VENDOR'S REPORTED SIZE DISTRIBUTION OF TUNGSTEN POWDERS

Micron size[a]	Fisher subsieve size, μ				
	0.88	1.15	2.1	3.5	4.5
	Percent by weight				
0-1	80.7	48.5	13.0	2.2	1.3
1-2	16.2	42.4	45.0	28.9	7.1
2-3	2.0	5.3	22.5	34.5	16.0
3-4	1.0	1.9	9.0	18.6	22.1
4-5	----	.8	6.5	9.1	22.7
5-6	----	.9	1.6	1.4	17.9
6-7	----	----	1.3	1.7	9.7
7-8	----	----	.7	1.0	1.2
8-9	----	----	----	1.1	2.1
9-10	----	----	----	1.3	.8

[a]By Photelometer.

The dispersed phase in the fuel plates was produced from high-fired (above 3000° F) UO_2 spheres depleted of the U-235 isotope and having a size range of -270/+400 mesh (about 53 to 37 μ). The chemical analysis for this material is given in table III. It was felt that the depleted UO_2 would be similar in character to the fully enriched UO_2, which would be used in the rocket reactor, since both types of UO_2 result from the same isotope separation process. Spherical UO_2 particles were used because irreg-

TABLE II. - VENDOR'S REPORTED CHEMICAL ANALYSIS OF 0.88-MICRON TUNGSTEN POWDER

Element	Parts per million (by weight)	Element	Parts per million (by weight)
Sodium	40	Nickel	<10
Potassium	60	Copper	10
Aluminum	<10	Manganese	<10
Calcium	20	Magnesium	<10
Silicon	<10	Tin	<10
Molybdenum	30	Carbon	20
Iron	100	Oxygen	1590
Chromium	10		

TABLE III. - VENDOR'S REPORTED CHEMICAL ANALYSIS OF URANIUM DIOXIDE

[Oxygen to uranium ratio, 2.00.]

Element	Parts per million (by weight)	Element	Parts per million (by weight)
Aluminum	150	Magnesium	<10
Boron	<.5	Manganese	<10
Carbon	19	Molybdenum	15
Calcium	<20	Nickel	<10
Cadmium	<.5	Lead	<2
Cobalt	<5	Silicon	<25
Chromium	<10	Tin	2
Copper	<6	Titanium	<10
Fluorine	10	Vanadium	<1
Iron	<20	Uranium	88.1 percent

ularly shaped particles might have sharp corners that could break off during fabrication and thus change the size distribution of the UO_2 in the fuel plates. The selection of the UO_2 size range used in this study was based on previous work (ref. 6) which indicated that fuel plates fabricated from this size range of UO_2 had the highest strength and best fuel-retention characteristics of any of those tested.

APPARATUS

The process used for fabricating W-UO_2 plates consists of blending, pressing, and sintering selected powders followed by hot rolling to attain high densities. The equipment used in each of these processes is described in the following paragraphs.

Blender

A twin-shell (V) blender operating at 25 revolutions per minute was used to mix the tungsten and UO_2 powders prior to pressing. The size of the powder lot determined the size of the blender to be used; for example, a 1-quart blender was used for a 1000-gram W-UO_2 lot.

Pressing Facilities

The powder was cold pressed at 20 000 pounds per square inch into a flat-plate configuration using the equipment described in references 4 and 5. A pressed plate size of 1 by 6 inches was used in this study.

Sintering Facilities

The pressed compacts were sintered in the hydrogen atmosphere tube furnace shown in figure 1. The heating element in this furnace is a 0.100-inch-diameter molybdenum wire, which is wound on a 24-inch-long, 3-inch-inside-diameter, high-purity alumina core. The core is positioned in a vacuum-tight, water-cooled, stainless-steel chamber, and the gap between the core and the

Figure 1. - Sintering furnace and controller.

chamber wall is filled with bubbled alumina insulation. A mechanical vacuum pump allows evacuation of the chamber prior to backfilling with commercial tank hydrogen to a positive pressure of approximately 3 pounds per square inch. A slight flow of hydrogen (approximately 1 cu ft/hr) is maintained through the furnace during the sintering cycle.

The power supply for the furnace is rated at 42 kilowatts and is controlled by an automatic temperature programer. The furnace temperature is monitored by a tungsten versus unalloyed rhenium thermocouple. The maximum furnace temperature is limited by the alumina core to about 3250° F.

Rolling Facilities

The sintered plates were hot rolled with an 8-inch, 4-high, rolling mill having 2.5-inch-diameter work rolls fabricated from an H12 tool steel. The backup rolls are 8 inches in diameter. The plates are heated to the rolling temperature in an induction furnace that is powered by a 10 000-cycle-per-second, 30-kilowatt motor generator. This furnace consists of a water-cooled copper induction coil that heats a 2-inch diameter tungsten susceptor. The 12-inch-long susceptor is made in 3-inch sections so that if one section fails, the entire susceptor does not have to be replaced. A high-purity alumina tube is used to insulate the susceptor from the copper coils while a molybdenum radiation shield between the susceptor and the alumina tube reduces the heat transfer to the alumina. The maximum furnace temperature is about 3800° F. The furnace is open at both ends, and a flat tungsten hearth extends through the furnace. Hydrogen is used as a furnace atmosphere and is allowed to burn at both ends of the furnace.

The furnace is placed directly in front of the rolling mill (as shown in fig. 2), and the samples are pushed through the furnace into the rolls. Transfer time from furnace to rolls is less than 1 second. A water-cooled runout table with a hydrogen atmosphere allows the samples to cool in a protective atmosphere.

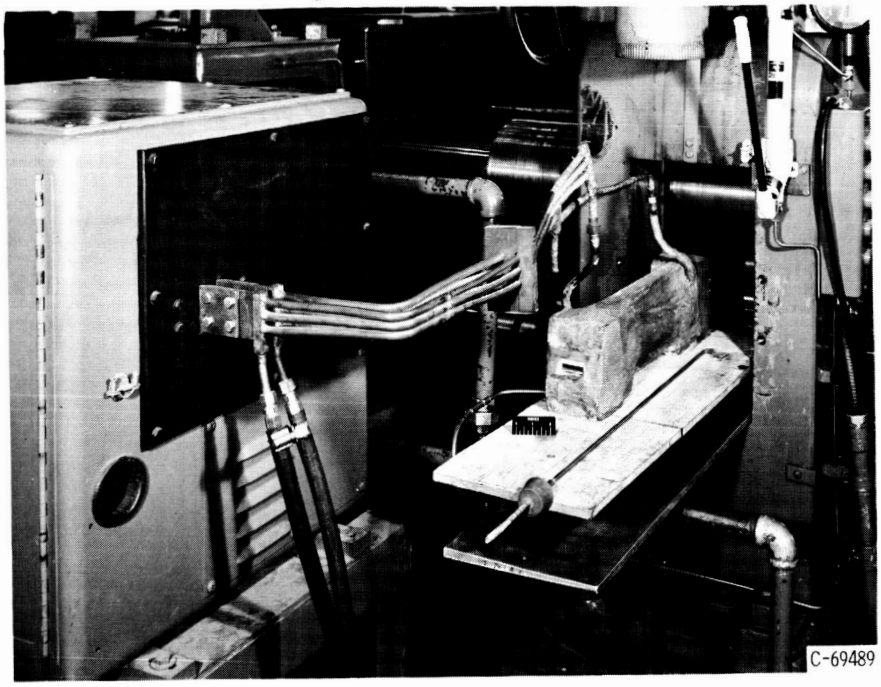


Figure 2. - Induction furnace and rolling mill.

DEVELOPMENT OF FABRICATION PROCEDURE FOR TUNGSTEN - 20-VOLUME-PERCENT URANIUM DIOXIDE FUEL PLATES

Previous work (refs. 4 and 5) demonstrated that it was feasible to produce W-UO_2 fuel plates by powder-metallurgy techniques. The particle size of the UO_2 (37 to 53 μ) used in this present study was much larger than the particle size of the UO_2 (3-μ average size) used in the previous fabrication studies; therefore, it was felt that modifications to the existing fabrication technique were necessary in order to optimize the fabrication procedure for the larger UO_2 particles. The existing fabrication technique, which consisted of blending, cold compacting, sintering, and hot rolling, was used as the starting point in this investigation.

Blending and Pressing

In the first step, the blending operation, weighed amounts of tungsten and UO_2 powders of the selected sizes were mixed together and blended in a twin-shell blender for 4 hours to give a homogeneous mixture. The relatively long blending time was necessary due to the poor blending characteristics of the coarse UO_2 particles and the fine tungsten particles. The mixture was then removed from the blender, and 2 percent (by weight) stearic acid was added to the mixture as both a die lubricant and binder. Acetone was used as a vehicle for the stearic acid. After thorough hand mixing of the W-UO_2 - stearic acid - acetone slurry, the acetone was evaporated from the mixture. The dried mixture was then broken up and passed through a 60 mesh screen. The stearic acid was subsequently vaporized from the composites during sintering, and no

carbon pickup from the stearic acid was noted in the sintered plates.

A weighed amount of the W-UO_2 mixture was hand loaded into steel dies and cold compacted at 20 000 pounds per square inch into a flat-plate configuration having a density of about 8.8 grams per cubic centimeter, which is approximately 50 percent of theoretical for the tungsten - 20-volume-percent uranium dioxide (W - 20 UO_2) plates. The compacts were dusted with zirconia and stacked into a tungsten sintering boat. Zirconia powder was used to prevent the compacts from sticking together during sintering.

Sintering

It was felt that compacts with high sintered densities could be rolled more readily than compacts of lower densities; therefore, a study was initiated to establish the sintering characteristics of the compacts with respect to tungsten particle size, temperature, and time.

Tungsten particle size. - In the study to determine the effect of tungsten particle size on the sintered density, unfueled tungsten plates were used and were fabricated in the same manner as W-UO_2 composites. Although the addition of UO_2 to the tungsten composite inhibited the sintering of the compact, as will be discussed later, it was assumed that all of the five tungsten powders used would be inhibited a similar amount. It was felt, therefore, that the tungsten particle size which gave the maximum sintered density for pure tungsten plates would also give the maximum density for the composite plates.

Five commercial tungsten powders, which had average particle sizes of 4.5, 3.5, 2.1, 1.15, and 0.88 microns, were used in this study. Two weight percent stearic acid was added to each lot of powder. The powder lots were cold pressed into plates and sintered in hydrogen at 3200° F for 20 hours. The very long-time sintering cycle minimized the effect of different sintering rates for the various tungsten powders since little additional sintering should occur after 20 hours at temperature.

The results of this study are given in table IV and in figure 3. Since the 0.88 micron tungsten powder sintered to the highest density (94.6 percent of theoretical) of the powders examined, this size was selected as the starting powder for the remainder of this investigation. Composite plates fabricated from the 0.88 micron tungsten powder and containing 20 volume percent UO_2 sintered to about 92.5 percent of theoretical density.

Sintering temperature. - It was shown in reference 4 that the sintered density of the W-UO_2 composites could be increased by increasing the sintering temperature. A tube furnace was constructed that had a higher temperature capability than the 2900° F furnace used in reference 5. The sintering temperature selected for this investigation was 3200° F because this temperature resulted in composites with high sintered densities and permitted relatively long furnace life. No observable UO_2 loss occurred in the W-UO_2 compacts held for 30 hours at this temperature under the slightly flowing hydrogen atmosphere.

TABLE IV. - PRESSED AND SINTERED DENSITIES OF COMPACTS

FABRICATED FROM VARIOUS SIZES OF TUNGSTEN POWDERS

[Sintering conditions, 20 hr at 3200° F in hydrogen.]

Average particle size, μ	Pressed density, percent theoretical	Sintered density, percent theoretical
0.88	----	94.2
.88	----	95.0
1.15	54.2	93.8
1.15	55.2	93.8
1.15	50.5	93.6
2.10	60.6	89.2
2.10	59.1	89.8
2.10	60.6	88.7
3.50	60.9	83.5
3.50	59.1	82.4
3.50	62.1	82.0
4.50	60.6	71.6
4.50	64.2	73.5
4.50	63.4	73.0

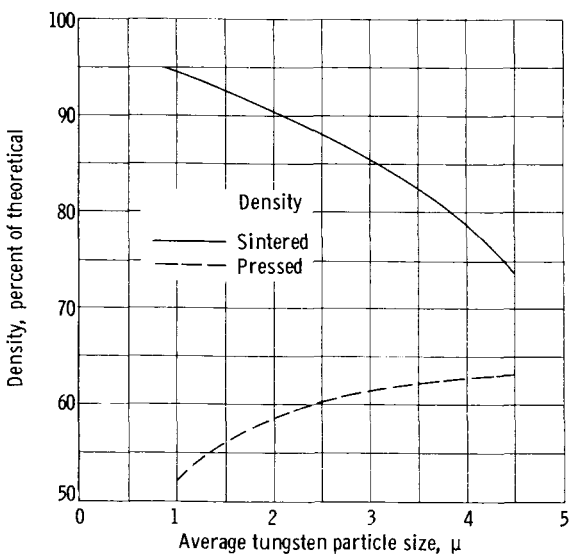


Figure 3. - Effect of initial tungsten particle size on both pressed and sintered densities.

Sintering time. - The effect of sintering time on the density of unfueled compacts was investigated to establish a sintering cycle. The compacts were fabricated from 0.88 micron tungsten powder and were sintered at 3200° F for 1, 4, 8, 15, and 30 hours. The results of this study are given in table V and figure 4 (p. 8). These results indicate that the sintered density increased with increasing sintering time up to about 8 hours. Although very little additional densification occurred after 8 hours, a sintering time of 15 hours was selected in this investigation for operating convenience.

Sintering cycle. - After the time and temperature parameters had been established for sintering the tungsten compacts, a sintering cycle was developed for the W-UO_2 composites.

This cycle is summarized as follows:

(1) The compacts were heated from room temperature to 700° F in 3 hours and held at this temperature for 1/2 hour to allow the stearic acid to vaporize from the compacts.

(2) The compacts were then heated to 2000° F in $2\frac{1}{2}$ hours and held at this

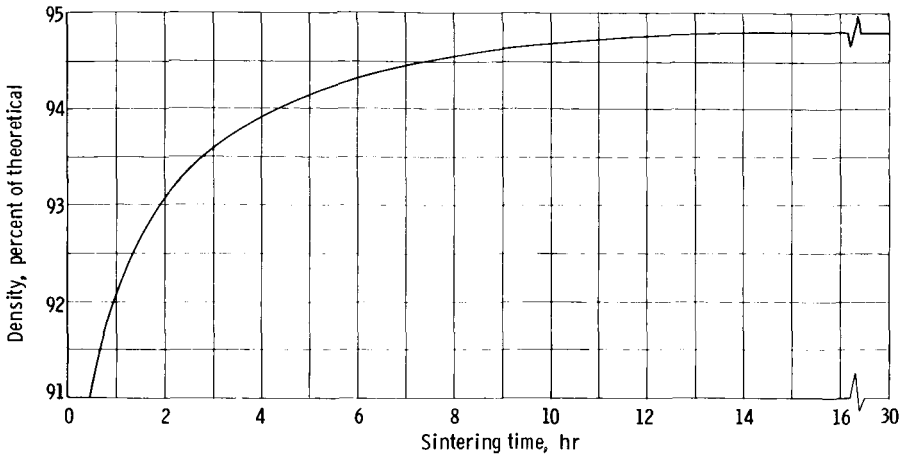


Figure 4. - Effect of sintering time on density of 0.88 micron tungsten (sintered at 3200° F in flowing hydrogen).

TABLE V. - SINTERED DENSITY OF TUNGSTEN COMPACTS AT VARIOUS TIMES

[Tungsten particle size, 0.88 μ; temperature, 3200° F.]

Time, hr	Density, percent theoretical
1	92.2
4	93.9
8	94.4
15	95.0
30	94.6

temperature for 1 hour to permit the hydrogen reduction of any surface oxide layers on the tungsten particles prior to sintering.

(3) The compacts were heated from 2000° to 3200° F in 2 hours and held at the sintering temperature for 15 hours.

(4) The compacts were then furnace cooled directly to room temperature in about 3 hours.

Cladding

The powder-metallurgy cladding method, described in reference 5, depends on cladding the compacts during the pressing operation. In this method, the W-UO_2 powder mixtures were sandwiched between layers of unfueled tungsten powder and subsequently pressed and sintered. This type of cladding was successful in retaining the UO_2, but the thickness and uniformity of the cladding were difficult to control. Therefore, a technique of cladding the composites by roll bonding wrought tungsten foil to the compacted and sintered W-UO_2 core was developed.

The surfaces of both the core and the clad must be clean for satisfactory roll bonding. For this reason, the major surfaces of the core were surface ground. Additional benefits gained by surface grinding include the removal of surface defects, the removal of any UO_2 depleted zones, and the removal of surface contaminations. The surfaces of the core and the thin (0.003-in.-thick) wrought tungsten foil cladding were scrubbed with acetone and dried. The foil was wrapped around the core as shown in figure 5 and fastened to the core at the tail end of the plate by spot brazing the cladding to the core with a small piece of tantalum foil. The brazed end of the plate passes through the rolls last, and thus contamination from the tantalum is minimized. The core-cladding

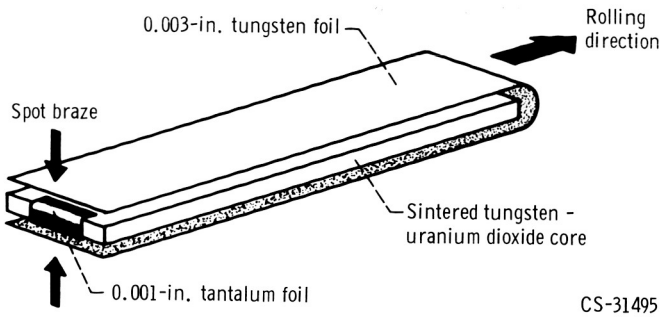


Figure 5. - Core-clad assembly of a fuel plate prior to rolling.

assembly is then hot rolled as will be discussed in the section on Rolling.

The clad produced, as a result of roll bonding, is of uniform thickness, fully dense, and metallurgically bonded to the core. Figure 6 shows a typical microstructure of the core-clad interface. Although there is porosity in the metal matrix, which is about 98-percent dense, there is no porosity in the fully dense clad. The clad thickness in this study, after rolling, was about 0.0015 inch; however, this thickness may be altered by changing the thickness of the starting foil and/or the amount of reduction during rolling. Fuel-retention tests have indicated that this cladding is very effective in preventing the loss of UO_2 from W-UO_2 fuel plates during isothermal elevated temperature tests (ref. 7).

Rolling

Preliminary hot-rolling studies at 4000° F on the W - 20 UO_2 composites fabricated in this investigation indicated that these plates possessed better rolling characteristics and exhibited much less edge cracking than composites containing the 3 micron UO_2 previously used. Thus, it was felt that it might be possible to roll this material at lower temperatures. A lower rolling

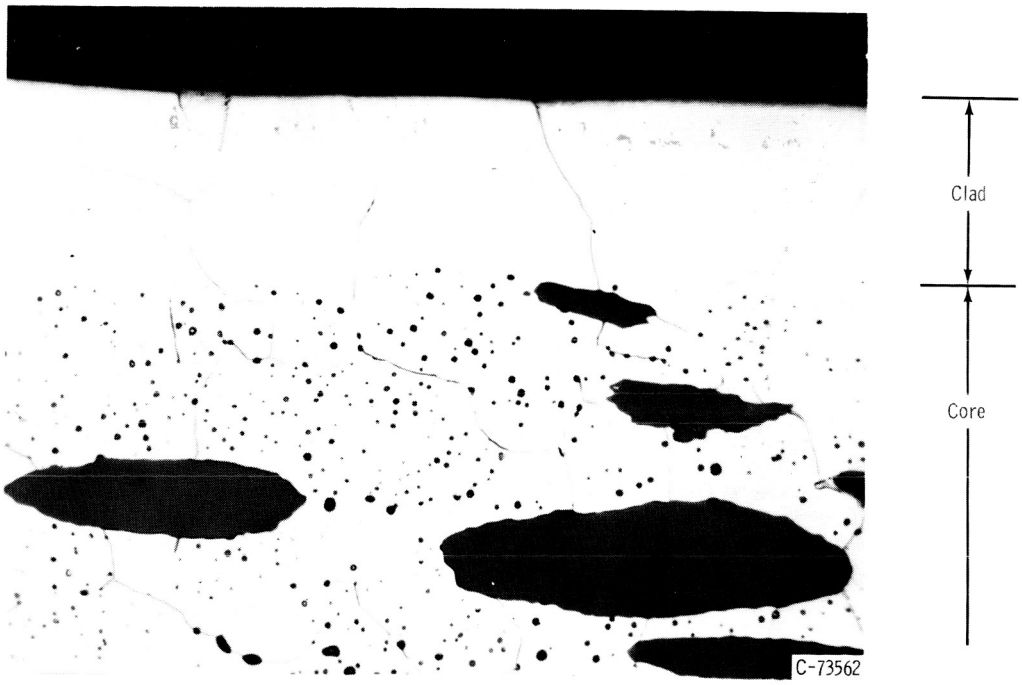


Figure 6. - Photomicrograph of core-clad interface after hot rolling. X500.

temperature would be beneficial since it would result in much longer rolling mill furnace life, and it would permit the use of alloy steel rolls in place of the more expensive molybdenum alloy rolls that were employed when rolling at 4000° F.

Rolling temperature. - The rolling temperature was lowered from 4000° F in order to find the minimum rolling temperature that would produce high quality plates with a minimum of edge cracking. This temperature was determined to be about 3550° F. At temperatures lower than this, the plate exhibited excessive edge cracking. During this study, the rolling speed was increased to about 200 surface feet per minute with a resulting decrease in the heat transfer from the plates to the work rolls. As a result of this decrease in heat transfer, the rolls stayed cooler and the effective rolling temperature of the plate was more nearly that of the rolling mill furnace.

Type of rolls. - As the rolling temperature was lowered, the hardness of the composites increased, but the hardness of the relatively soft molybdenum alloy rolls remained constant. Below about 3800° F, the composites began to mark the rolls. At 3550° F, only a few passes could be made before the roll surfaces began to deteriorate, and the surface finish of the composites became unacceptable. Work rolls of several other compositions were tried, and the most satisfactory results were obtained with H12 tool steel rolls hardened to 53 to 54 Rockwell-C.

The hot composites had a tendency to stick or weld to the clean alloy steel rolls. This problem was solved by conditioning the roll surfaces by rolling unalloyed tungsten sheet at about 3100° F. After a thin oxide layer had been built up on the rolls, the W-UO_2 composites could be satisfactorily rolled at 3550° F.

Rolling schedule. - The rolling schedule, which was developed for roll cladding the compacts and for rolling them to final thickness, consisted of a 5-percent reduction in area on the initial pass, followed by a 20-percent reduction on the second pass. A 10-percent reduction was taken on all subsequent passes until a total overall reduction of between 40 and 50 percent was achieved. The end of the plate, which was contaminated with the tantalum brazing foil, was cut off after the second pass.

EFFECT OF HOT WORK ON PROPERTIES OF TUNGSTEN - 20-VOLUME-PERCENT URANIUM DIOXIDE COMPOSITES

The effect of hot work was studied to determine what effect this variable would have on the final properties of the fuel plates. Compacts containing 20 volume percent UO_2 were pressed and sintered using the procedure previously described. The thickness of the compacts was varied by loading different amounts of the W-UO_2 powder mixture into the pressing dies. The compacts were then hot rolled at 3550° F to the same final thickness, and the resultant amount of hot work in the rolled plates, as measured by reduction in area, varied from 0 to 80 percent. The density, high temperature strength, and bend

TABLE VI. - EFFECT OF HOT WORK ON DENSITY, MECHANICAL PROPERTIES, AND 4t BEND TRANSITION TEMPERATURE OF TUNGSTEN - 20-VOLUME-PERCENT URANIUM DIOXIDE COMPOSITES

Hot work, percent reduction	Density, percent theoretical	Ultimate tensile strength at 4500° F, psi	Yield strength at 4500° F, psi	Bend transition temperature, °F
0	92.0	2470	2160	>900
0	----	2750	2440	----
10	94.7	3640	3450	>900
10	----	3500	3020	----
21	95.4	3560	3380	>900
21	----	3820	3710	----
21	----	3630	3560	----
31	96.1	4160	3960	680
41	97.9	4000	3920	775
41	----	3860	3800	----
41	----	3810	3775	----
51	98.4	----	----	580
59	99.3	4370	3860	680
59	----	3690	3560	----
70	99.6	----	----	----
80	99.8	3870	3400	>900
80	----	3660	3330	----
80	----	3790	3420	----

ductility of the plates were measured, and the microstructures of the rolled plates were examined.

Density and Microstructure

The densities of the rolled plates, as measured by a water displacement method, are listed in table VI and plotted as a function of the amount of hot work in figure 7. These data indicate that the density of the rolled plates increases as the amount of hot work increases. In order for the density of the worked compacts to exceed 98 percent of theoretical density, the sintered compact (92.5 percent of theoretical density) must be worked more than 40 percent.

The changes in microstructure as a result of hot working are shown in the photomicrographs of figure 7. Small voids are present in the tungsten matrix of the as-sintered composite. These small voids are believed to result from the fully dense UO_2 particles blocking or inhibiting the sintering of the tungsten matrix. Hot work is believed to promote the closing of these voids. The photomicrographs of worked structures in figure 7 show no evidence of voids, even after a reduction of only 20 percent in area. In the microstructure of the 20-percent-worked sample, however, there is still evidence of fine porosity in the tungsten matrix, but as the amount of hot working increased, the amount of porosity in the matrix apparently decreased. In the 60-percent-worked sample, which is over 99-percent dense, there is no visible porosity at a

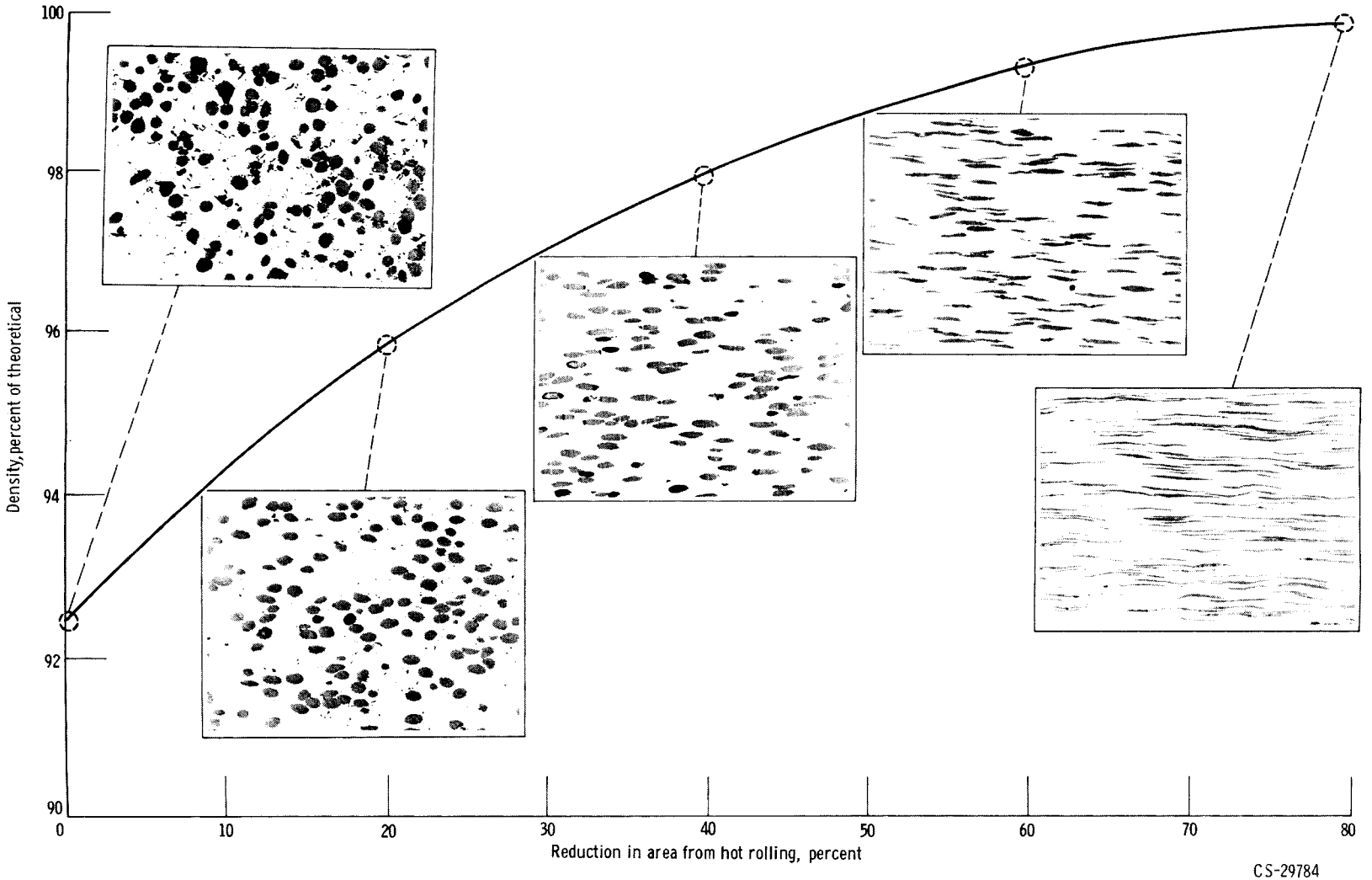
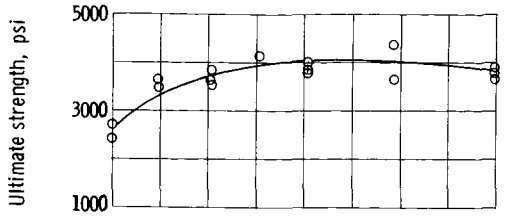
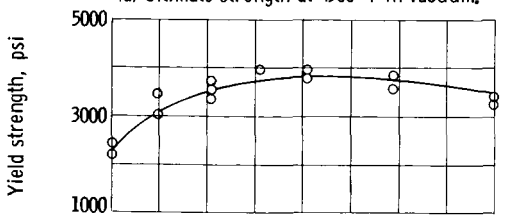


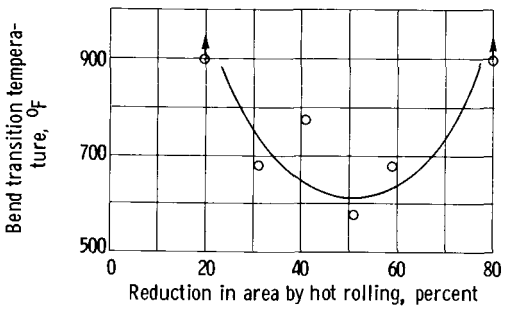
Figure 7. - Effect of hot work on density and microstructure of tungsten - 20-volume-percent uranium dioxide plates. X100. (Reduced 50 percent in printing.)



(a) Ultimate strength at 4500° F in vacuum.



(b) Yield strength at 4500° F in vacuum.



(c) Bend transition temperature for 4t radius.

Figure 8. - Effects of hot work on ultimate tensile strength, yield strength, and 4t bend transition temperature of tungsten - 20-volume-percent uranium dioxide plates.

magnification of X100.

In addition, as the amount of working increased, the UO_2 particles in the composites deformed and elongated. The UO_2 in the as-sintered material is nearly spherical, while the UO_2 in the 80-percent-worked sample is highly stringered, indicating that the UO_2 is plastic at the rolling temperature used.

Tensile Strength and Bend Ductility

The ultimate tensile strength, yield strength, and 4t bend transition temperature are listed in table VI and are plotted as a function of the amount of hot work in figure 8. The longitudinal axes of all the test specimens were parallel to the rolling direction.

The tensile tests were conducted in a vacuum of 5×10^{-5} torr or better at 4500° F using the procedure described in reference 8. As expected, both the ultimate tensile strength and the yield strength increased with working up to about 40-percent work. This increase in strength is believed to be due to the increase in the density of the matrix and the subsequent elimination of porosity. Very little additional change was observed in the strength of material worked more than 50 percent.

The ductile-to-brittle bend transition temperature was measured (in air) as a function of the amount of hot work. In this series of tests, the bend radius was four times the thickness of the test specimens. Somewhat surprisingly, a minimum transition temperature (approximately 600° F) occurred at about 50-percent work. The initial decrease in transition temperature with increasing work is again believed to be due to an increase in the density of the matrix. The reason for the increase in transition temperature for the material worked more than 50 percent is not fully understood; however, one possible explanation is that there is a higher probability of interconnection of the elongated UO_2 particles with increasing work. It is felt that this interconnection results in a more brittle structure.

Rolling Characteristic

Gross observations on the rolling characteristics of the plates indicated that very little edge cracking occurred up to about 50-percent hot work; how-

TABLE VII. - EFFECT OF SINTERED DENSITY OF TUNGSTEN - 20-VOLUME PERCENT URANIUM DIOXIDE COMPOSITES ON ROLLED DENSITY AND ROLLING CHARACTERISTICS

Sintered density, percent theoretical	Rolled density, percent theoretical	Rolling characteristics
64.5	95.3	Very poor
75.3	96.2	Poor
80.9	95.3	Poor
86.0	97.0	Fair
92.5	98.4	Good

ever, at 60-percent work, edge cracking became more severe. The plates with 70- and 80-percent work were very badly edge cracked, and most of these plates had to be scrapped.

Conclusions

Although the maximum strength and minimum bend transition temperature were obtained on plates that were rolled about 50 percent, results of fuel loss tests at 4500° F on specimens rolled various amounts indicated that plates reduced about 50 percent lost fuel at a faster rate than plates worked a lesser amount. The higher fuel loss rate from the more heavily worked specimens was probably also due to more interconnection of the UO_2 particles. A compromise between strength and fuel retention was therefore necessary. The amount of reduction selected for the standard fabrication technique was such that the density of the rolled plates exceeded 98 percent of theoretical density. This amount of work corresponds to about 42-percent hot work for the fuel plates under discussion.

EFFECT OF SINTERED DENSITY ON ROLLING CHARACTERISTICS

A study was made to determine if high sintered densities were required for good rolling characteristics and, if high densities were required, to determine the minimum density that would give good results. Unclad compacts containing 20 volume percent UO_2 were sintered at various temperatures such that the sintered densities were 64.5, 75.3, 80.9, 86.0, and 92.5 percent of theoretical. These compacts were then hot rolled at 3550° F to a total reduction of 50 percent. The results of this study are summarized in table VII and are shown in figure 9. These data indicate that a sintered density of about 91 percent of theoretical was required before the rolled density exceeded 98 percent of theoretical at 50-percent hot work.

The compacts that were under 86 percent of theoretical density did not roll well, and numerous small cracks appeared on the major surfaces of the rolled plates. The effect of sintered density on the surface appearance of the rolled plates is shown in figure 10. Horizontal cracks on the surfaces of the plates are shown in figures 10 (a), (b), and (c). As the sintered density

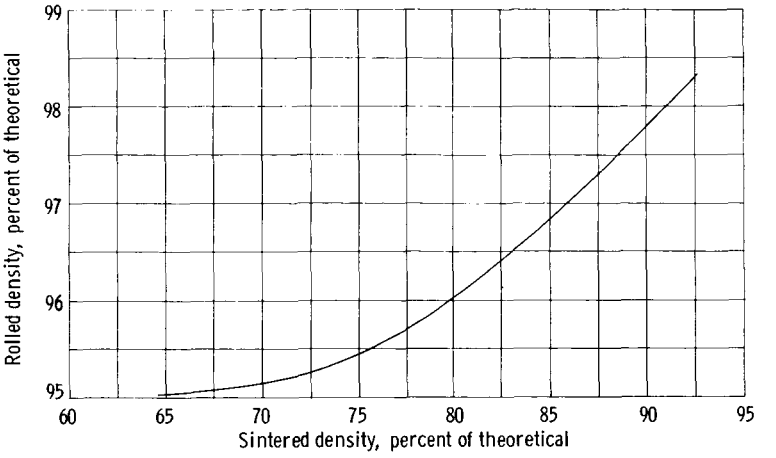


Figure 9. - Effect of sintered density on rolled density for 50-percent hot-worked tungsten - 20-volume-percent uranium dioxide.

increased, both the number and size of the surface cracks decreased. The compact that had a sintered density of 64.5 percent of theoretical cracked apart after only two reductions (28-percent total reduction in area).

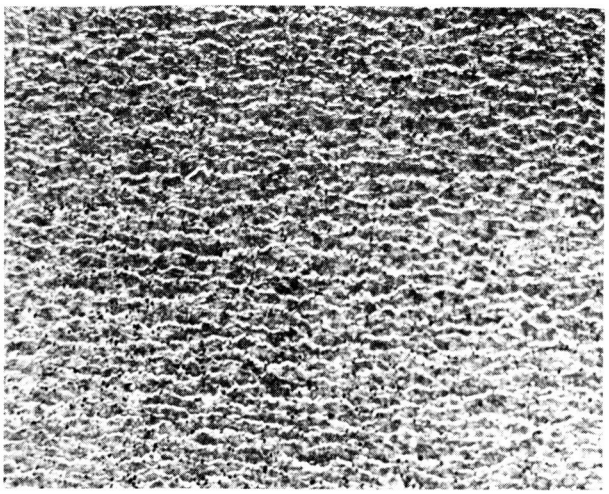
The compacts that were 86.0- and 92.5-percent dense rolled in a similar manner except that the denser compact exhibited slightly less edge and surface cracking. The denser compact also rolled to a higher final density.

These results indicate that high-density compacts are more desirable since they rolled better than the low-density compacts and since they permitted a higher rolled density to be obtained. A minimum density of about 86 percent of theoretical is required for satisfactory rolling of composites of this type.

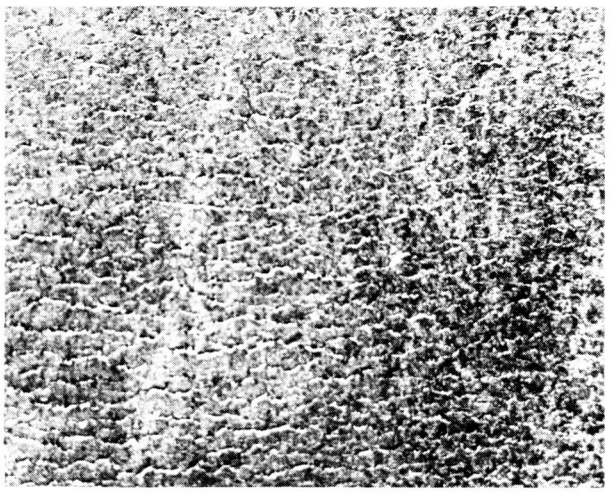
EFFECT OF FUEL LOADING ON FABRICATION OF FLAT PLATES

A study was made to determine if the fabrication procedure developed for the W-UO_2 fuel plates containing 20 volume percent UO_2 could be employed to fabricate fuel plates containing 10 to 40 volume percent UO_2. Compacts containing 0 to 40 volume percent UO_2 were processed using the fabrication procedure described in this report. The compacts were roll clad and a total overall reduction by rolling of 40 percent was employed. The compacts were then evaluated with respect to sintered density, rolling characteristics, and microstructure. The results are summarized in table VIII and figures 11 and 12. As was noted previously, the addition of UO_2 to the tungsten inhibited the sintering of the compacts. Evidence of this impaired sintering is shown in the plot of sintered density as a function of the fuel loading of W-UO_2 composites in figure 11. For this particular fabrication technique, the densities (percent theoretical) of the compacts decreased as the amount of fuel in the compacts increased.

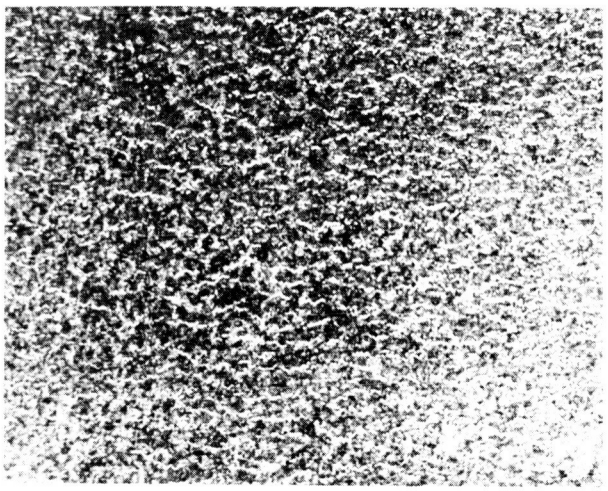
No problems were encountered in hot rolling the compacts containing up to 40 volume percent UO_2. There was, however, a greater tendency for edge cracking as the fuel loading increased. The roll-bonding technique worked well with



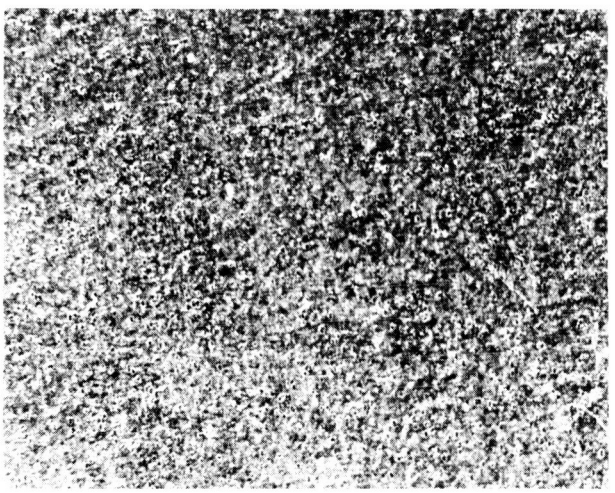
(a) Sintered density, 64.5 percent.



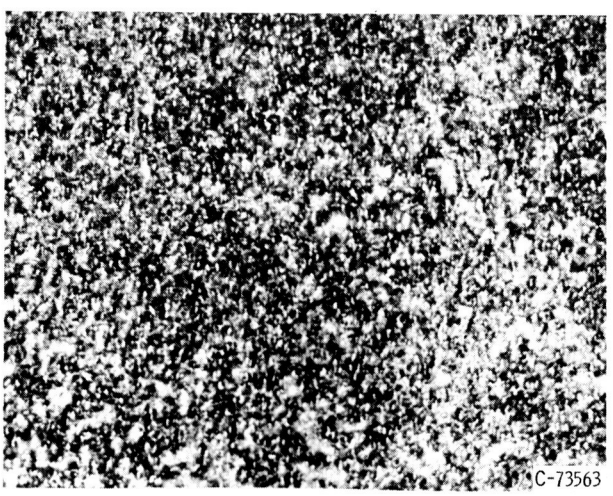
(b) Sintered density, 75.3 percent.



(c) Sintered density, 80.9 percent.



(d) Sintered density, 86.0 percent.



(e) Sintered density, 92.5 percent.

Figure 10. - Effect of sintered density on surface finish of hot-rolled tungsten - 20-volume-percent uranium dioxide composites. X10.

TABLE VIII. - EFFECT OF FUEL LOADING ON SINTERED DENSITY AND ROLLING CHARACTERISTICS OF TUNGSTEN - URANIUM DIOXIDE COMPOSITES

Fuel loading, volume percent uranium dioxide	Sintered density, percent theoretical	Rolling characteristics
0	94.6	Good
5	94.0	
10	92.4	
10	95.3	
15	91.7	
20	93.0	
20	92.0	
30	88.8	
40	83.3	Fair (edge cracking)

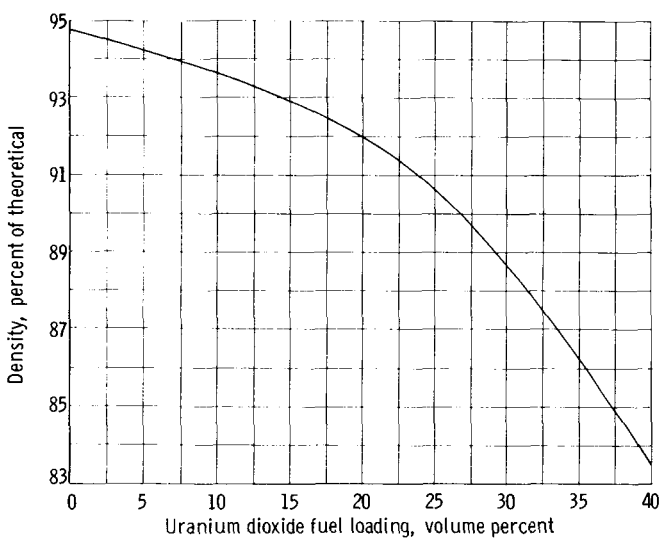


Figure 11. - Effect of uranium dioxide fuel loading on sintered density of tungsten - uranium dioxide compacts.

all fuel loadings and no problems were encountered with bonding defects.

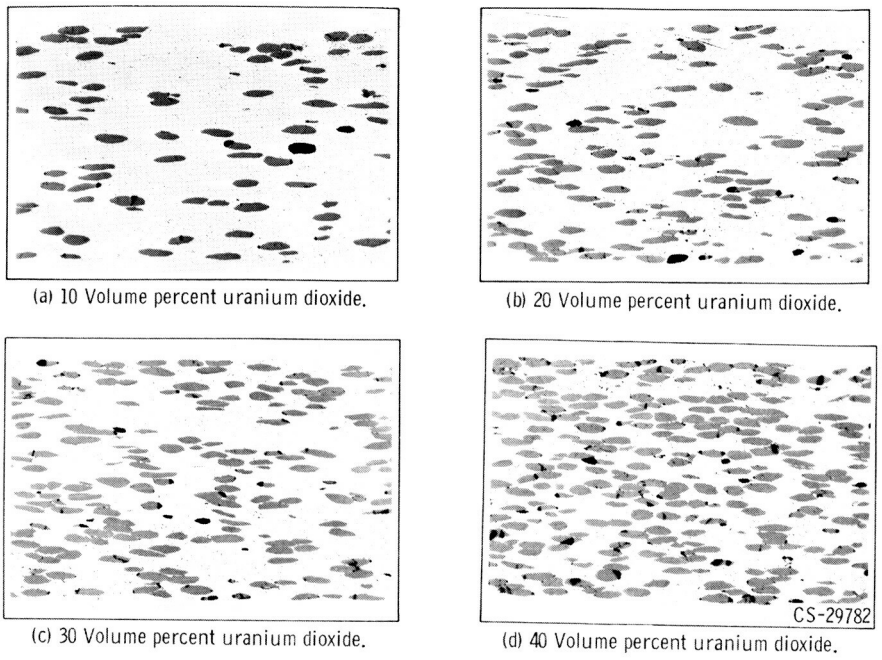
Photomicrographs of longitudinal sections of four specimens containing different fuel loading are shown in figure 12. These samples were all hot worked a similar amount (40 percent). The density of the core, as indicated by the microporosity in the tungsten, tended to decrease as the fuel loading increased. This is because the higher fuel loadings did not sinter as well, and thus, at a given amount of hot work, composites with higher fuel loadings were less dense.

The results indicate that the addition of UO_2 to the tungsten matrix impairs the sintering of the compacts. It is possible, however, to fabricate fuel plates containing up to 40 volume percent UO_2 by the method described in this report.

FORMING OF TUNGSTEN - URANIUM DIOXIDE FLAT PLATES INTO OTHER CONFIGURATIONS

Since one of the fuel-element designs under consideration at the Lewis Research Center employs thin-walled concentric cylinders of W-UO_2, an attempt was made to form flat fuel plates containing 20 volume percent UO_2 into cylinders. This material had a bend transition temperature of less than 700° F. Since the bend transition temperature of a material represents the temperature at which the material will bend under optimum conditions (i.e., uniform heating and hot dies), the actual forming temperature was expected to be somewhat higher.

A series of concentric rings was successfully formed by heating the fuel plates to about 1700° F and then shaping them around a mandrel. Figure 13 shows the results of this forming operation. The smallest cylinder shown is about 1/2 inch in diameter, and the largest cylinder is about 1 inch in dia-



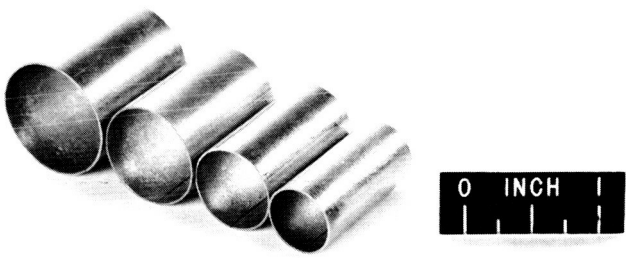
(a) 10 Volume percent uranium dioxide.

(b) 20 Volume percent uranium dioxide.

(c) 30 Volume percent uranium dioxide.

(d) 40 Volume percent uranium dioxide.

Figure 12. - Effect of fuel loading on microstructure of tungsten - uranium dioxide compacts. X100. (Reduced 50 percent in printing.)



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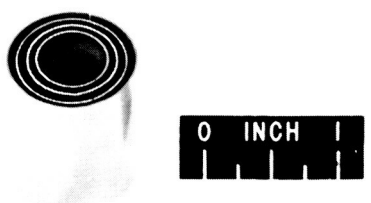

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Figure 13. - Cylinders hot formed from tungsten - 20-volume-percent uranium dioxide plates.

meter. The axes of these cylinders are perpendicular to the rolling direction in the original plates. It was not possible to form a cylinder with the axis parallel to the rolling direction. When the plates were bent in this direction they fractured. This is probably due to the elongated fuel particles and the probable resultant anisotropy.

Cylinders were also formed from plates containing 10 and 30 volume percent UO_2, but plates containing 40 volume percent UO_2 cracked during forming. The photomicrograph of the fuel plate containing 40 volume percent UO_2 (fig. 12) shows that there is almost a continuous path of brittle UO_2 through the tungsten matrix, which could lead to brittle fractures during forming.

An attempt was also made to form a corrugated shape from plates containing 20 volume per-

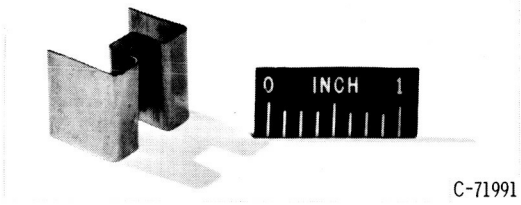

Figure 14. - Corrugated shape hot formed from a tungsten - 20-volume-percent uranium dioxide plate.

cent UO_2. The plate was hot formed at about 1700° F into the configuration shown in figure 14. Each of the corrugations was about 1/4 inch on a side and a bend radius of less than 1/32 of an inch was achieved at all bends. The results of this attempt show that it is possible to form relatively complex shapes from W-UO_2 plates.

One lot of fuel plates containing 20 volume percent UO_2 could not be bent without cracking (even at 2500° F). A particle size analysis of the starting UO_2 powders showed that about 2 percent of the UO_2 was less than 20 microns in diameter. However, plates that were fabricated from this same lot of UO_2 powder but with the minus 20 micron fraction removed could be formed without difficulty. These results indicate that the formability of the fuel plate is decreased by the presence of fine UO_2 particles.

SUMMARY OF RESULTS

From this study of the fabrication of W-UO_2 fuel plates containing UO_2 in the size range of 37 to 53 microns, the following conclusions have been reached:

1. It is possible to fabricate dense clad W-UO_2 plates containing 10 to 40 volume percent UO_2 by the procedure now summarized:

   a. Blend weighed amounts of tungsten and UO_2 powders in a V-blender for 4 hours.

   b. Add 2 weight percent stearic acid in acetone vehicle to blend. Dry the mixture and pass through a 60 mesh screen.

   c. Press at 20 000 pounds per square inch.

   d. Sinter in flowing hydrogen for 15 hours at 3200° F.

   e. Surface grind major surfaces of sintered plates and attach cladding.

   f. Roll at 3550° F, using a rolling schedule of 5 percent reduction on the first pass, 20 percent reduction on the second pass, and 10 percent reduction on all subsequent passes. An overall reduction of 40 to 50 percent is required.

2. The major surfaces of the fuel plates can be clad with a thin, dense, uniform layer of pure tungsten by hot roll bonding. The resultant cladding is metallurgically bonded to the core.

3. The UO_2 in the composites flows plastically as the compact is hot

worked at temperatures near 3600° F. The strength and bend ductility of fuel plates containing 20 volume percent UO_2 increase with working up to about 50-percent work, but severe working increases the fuel losses from the compacts at elevated temperatures. Therefore, a compromise is necessary. For this reason, an amount of hot work that provides a density of 98 percent of theoretical is selected for this study. This corresponds to about 42 percent work for the W - 20 UO_2 fuel plates.

4. For successful fabrication by hot rolling, using the method described, a sintered density of a least 86 percent of theoretical is required.

5. The W-UO_2 fuel plates can be hot formed at 1700° F into both simple cylinders and complex shapes.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, December 17, 1964.

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